

Glassy behavior of high- T_c superconductors

by I. Morgenstern

This paper deals with the question of flux creep or glassy behavior in high- T_c superconducting single crystals. It is shown that the flux creep picture is merely a phenomenological approach to the glassy behavior for relatively short times and low temperatures. Glassy effects are predicted for temperatures between 70% and 95% T_c and magnetic fields in the range of 0.03 T to 0.2 T. The glass concept can be understood as a generalization of the traditional flux creep picture. A hierarchy of energy barriers dominates the physical behavior. An important technical aspect is the influence of the glassiness on critical currents.

Introduction

At the beginning we want to clarify that in the case of the macroscopic glass picture, we consider glassy behavior inside a single grain or single crystal, and not in a ceramic. **Figure 1** shows the situation. On level 1 we have a disordered array of grains, a ceramic. Inside a grain we see the macroscopic glass consisting of weakly coupled two-dimensional (2D) planes. Each plane (level 3) consists of an array of domains, probably due to the heavy twinning of the system, but other defects may also cause these domains (e.g., oxygen deficiencies). The resulting weak links are essential for the glassy behavior. Level 4 inside the domain describes the

microscopic mechanism. It should be noted that the macroscopic glassy behavior is not the origin of high- T_c superconductivity, and therefore we do not consider level 4.

The first experiment stating glassy behavior in high- T_c materials was carried out by Müller et al. [1(b)]. In the spirit of spin-glass experiments [2, 3], they considered zero-field cooling (z.f.c.) versus field-cooling (f.c.) behavior for a ceramic La-Ba-Cu-O system. Measuring the susceptibility (**Figure 2**), they found reversible behavior after field cooling. In sharp contrast, metastability showed up in the irreversible behavior after z.f.c. The two curves met at a temperature further on denoted as $T_c^*(H)$ (H = magnetic field), above which only reversible behavior was detected. Thus, $T_c^*(H)$ is the temperature below which metastable behavior occurs, just as in the corresponding spin-glass experiment.

Measuring in different magnetic fields clarified the analogy to spin glasses even more. A quasi-de-Almeida-Thouless line [4] was found; i.e., we have

$$H^{2/3} \sim T_c^*(0) - T_c^*(H), \quad (1)$$

just as in spin glasses (**Figure 3**). The prefix "quasi" had to be added, as further theoretical work [5] showed that the underlying mechanism for the $H^{2/3}$ behavior is different from that in spin glasses.

Furthermore, experiments [1(b)] also showed nonexponential decay of magnetization or susceptibility (insert of **Figure 2**) also reminiscent of spin glasses. Therefore, a natural theoretical approach to the problem had to consider various (spin) glass models. Carrying out numerical simulations for an XY spin-glass model based on earlier work of Ebner and Stroud [6], we were able to repeat the experimental findings. **Figure 4** shows the f.c. and z.f.c. susceptibilities for various magnetic fields H . The resulting error bars denote $T_c^*(H)$. **Figure 5** shows nothing

©Copyright 1989 by International Business Machines Corporation. Copying in printed form for private use is permitted without payment of royalty provided that (1) each reproduction is done without alteration and (2) the *Journal* reference and IBM copyright notice are included on the first page. The title and abstract, but no other portions, of this paper may be copied or distributed royalty free without further permission by computer-based and other information-service systems. Permission to republish any other portion of this paper must be obtained from the Editor.

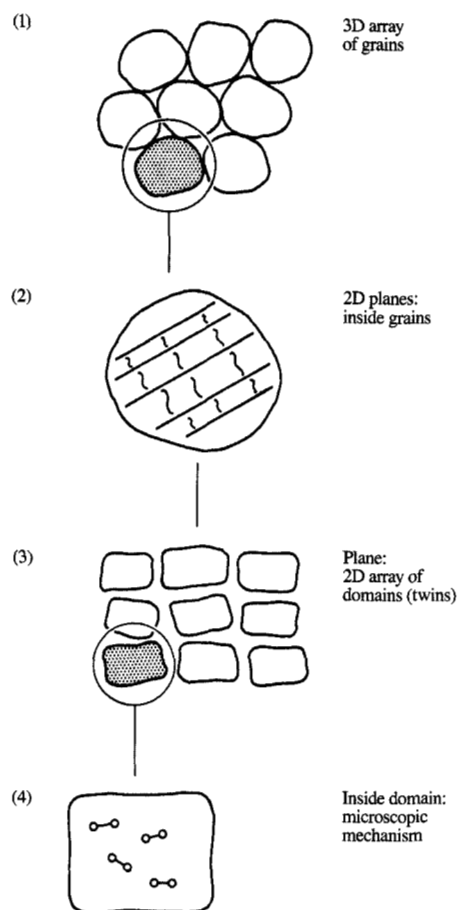


Figure 1

Schematic display of the physical situation in a ceramic (single-crystal level 2). From [1(a)].

unexpected: plotting $H^{2/3}$ as a function of $T_c^*(0) - T_c^*(H)$ results in a straight line. Furthermore, the model was surprisingly successful in describing the features of high- T_c glass experiments [7]. We refer the reader to [5] for details.

The theoretical model of the XY spin glass of Ebner and Stroud [6] can be understood as a disordered array of Josephson junctions. The following Hamiltonian describes the system:

$$-\beta \mathcal{H} = J \sum_{\langle ij \rangle} \cos(\phi_i - \phi_j - A_{ij}). \quad (2)$$

The ϕ_i (the XY spins) describe the regions or domains of coherent phases in the system, originally the (physical) grains of the ceramic or the granular superconductor, as considered

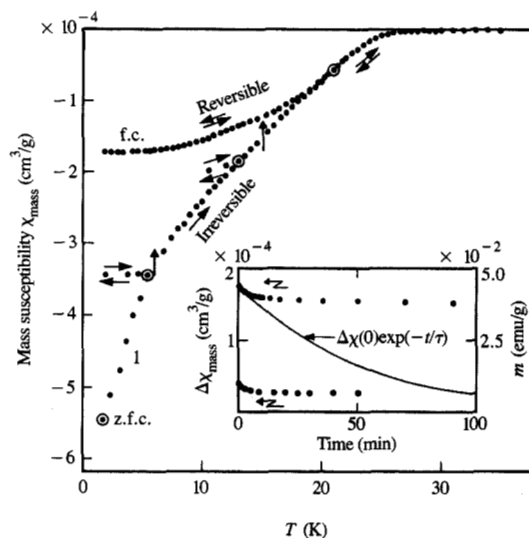


Figure 2

Susceptibility versus temperature. Experimental situation for La-Ba-Cu-O. Field-cooling and zero-field-cooling measurements. Insert: nonexponential time decay. From [1(b)].

in [6]. However, it soon became clear that our La-Ba-Cu-O ceramic behaved differently—the domains of the glass model had to be located *inside* the grains. Deutscher and Müller [8] solved the resulting puzzle. The extremely short coherence length in the system and the existence of twin boundaries lead to Josephson junctions or, more generally, weak links inside the grains. In general, however, we may consider the XY spin-glass model as an approximation to the Landau-Ginzburg theory omitting higher-order terms but including disorder. The resulting disorder is described by the phase factor A_{ij} in the model which is given by the line integral

$$A_{ij} \sim \int_i^j \vec{A} \cdot d\vec{l} \quad (3)$$

from site i to site j over the vector potential $\vec{A}$. The sites i and j reflect the positional disorder in the system.

The main feature of Hamiltonian (2) is frustration. Figure 6 shows the effect well known to spin-glass physicists [2]. Spin 4 just does not know whether to follow spin 1 according to phase factor A_{41} or spin 3 due to A_{34} . It is frustrated. The origin of frustration is disorder leading to "randomized" A_{ij} . The interaction in the system via the weak links leads to cooperative effects—the glassy behavior. Glassy behavior is best described by considering the energy landscape of the system (Figure 7). Again, it is well known that we have a hierarchy of barriers, as shown in the 2D cut

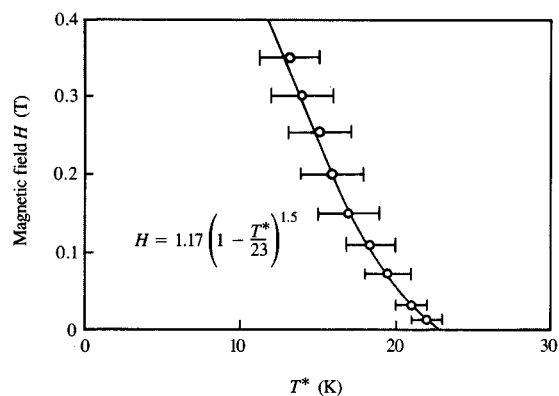


Figure 3

Quasi-de-Almeida-Thouless line experiment. From [1(b)].

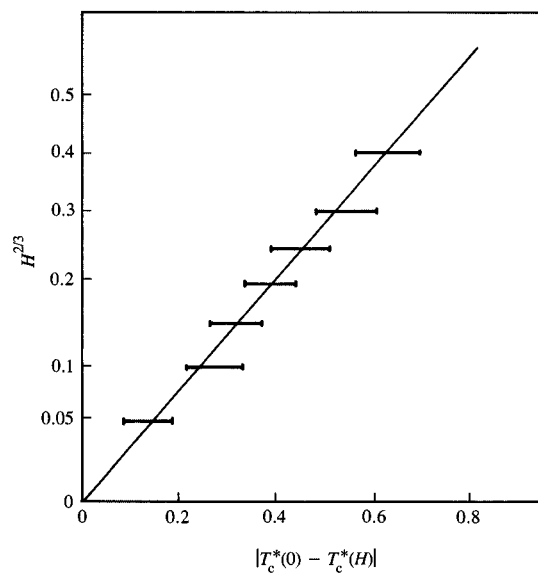


Figure 5

Quasi-de-Almeida-Thouless line; numerical simulation. From [5].

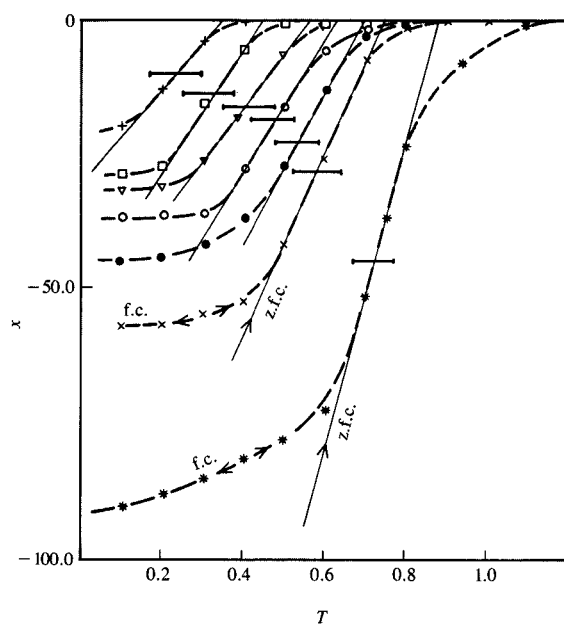


Figure 4

Susceptibility versus temperature; numerical simulation; various magnetic fields from $H = 0.05$ (lowest curve) to $H = 0.40$ in steps of $\Delta H = 0.05$ in units of $2\pi/\phi_0$ (except $H = 0.35$). ϕ_0 is the elementary flux quantum. From [5].

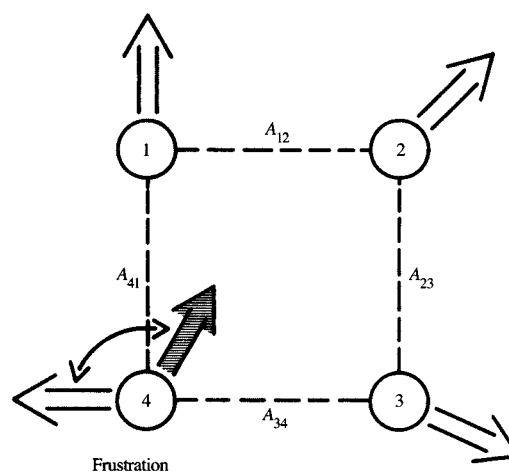


Figure 6

Frustration in theoretical model (see text). From [5].

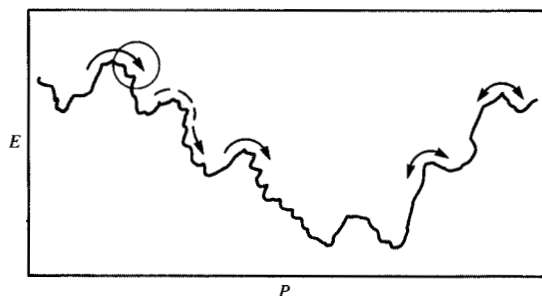


Figure 7

Energy landscape of glassy system. Hierarchy of barriers. From [21].

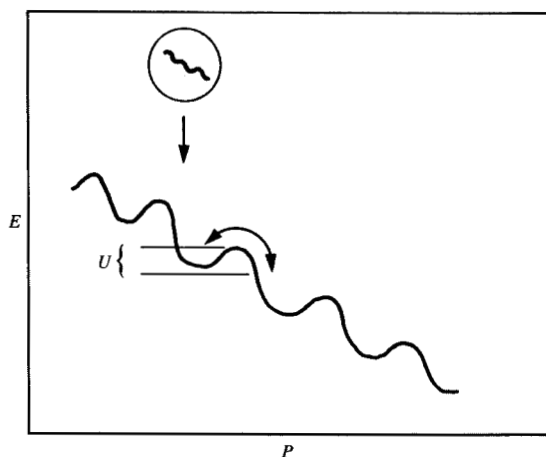


Figure 8

Enlarged slope of energy hill. Corresponds to giant flux creep picture. From [21].

through the multidimensional phase space (denoted by coordinate P). Most of the behavior results from the fact that the system has to hop over not only the small barriers, but also the larger ones, i.e., the complete hierarchy of barriers in the system.

Mota et al. [9] realized that the decay of the magnetization with time showed $M(t) \sim \ln t$. They compared their result with the traditional flux creep picture, which indeed predicted this type of decay. Malozemoff et al. [10] even went a step further: They argued that the traditional flux

creep picture [11] led to a sufficient description of their experimental findings, considering just a single or only a few barriers originating from the pinning forces acting on the flux lines. It is important to note that these experiments were carried out at low temperatures and only "relatively" short observation times. In the work of Mota et al. [9], the measurements were taken over a period of 24 hours, but at the "relatively" low temperatures (about helium temperature) no deviation from the $\ln t$ behavior was found. Here it has to be noted that the emphasis is on the term "relatively," as we shall see in the next paragraph.

Back to Figure 7, our hierarchy of barriers. Just considering the slope of a hill, we know from spin-glass research that this slope consists of a "rough surface" [12], as shown in Figure 8. It immediately becomes clear that the flux creep theory, based on a similar phenomenological physical picture, describes only the decay of the system one single slope down. Considering the average size of a barrier U , the experiments of Malozemoff et al. [10] show that it is (a) relatively too low for the traditional flux creep picture, but (b) also relatively too high for the glass picture, again in the traditional sense. Malozemoff's reasoning leads to the introduction of the giant flux creep picture, just allowing more flux to creep over the relatively small barriers [10]. But what happens at higher temperatures, when the hopping probabilities are higher? Is the system then able to experience our nice hierarchy of barriers? The answer is certainly yes! Giant flux creep or the influence of only a few barriers is seen at temperatures below about 70% T_c and for relatively short times which can be up to several days or weeks at helium temperatures. Here the phenomenological approach to the problem, the flux creep picture, is certainly valid; yet we must always keep in mind that we are dealing with a glass, but with such large relaxation times that they do not show up in ordinary low-temperature experiments. Above about 70% T_c , however, glassy behavior can be detected on relatively reasonable time scales (Figure 9). The appropriate magnetic fields are roughly in the range of 0.03 T to 0.2 T. Too close an approach to T_c destroys the glassy behavior again, as we experience critical effects. Magnetic fields outside the above range take the system out of the glass regime for the temperatures mentioned [5].

The general message of this paper is the following: In the regime of about 70%–95% T_c , we will find glassy behavior and a new rich physics in the field of superconductivity. What do we have to expect and what is already known?

1. Kohlrausch decay

The magnetization $M(t)$ should decay as

$$M(t) \sim \frac{e^{-(t/\tau)^\beta}}{t^x}, \quad (4)$$

with $0 < \beta < 1$ and $0 < x < 1$; τ is the relaxation time. Note

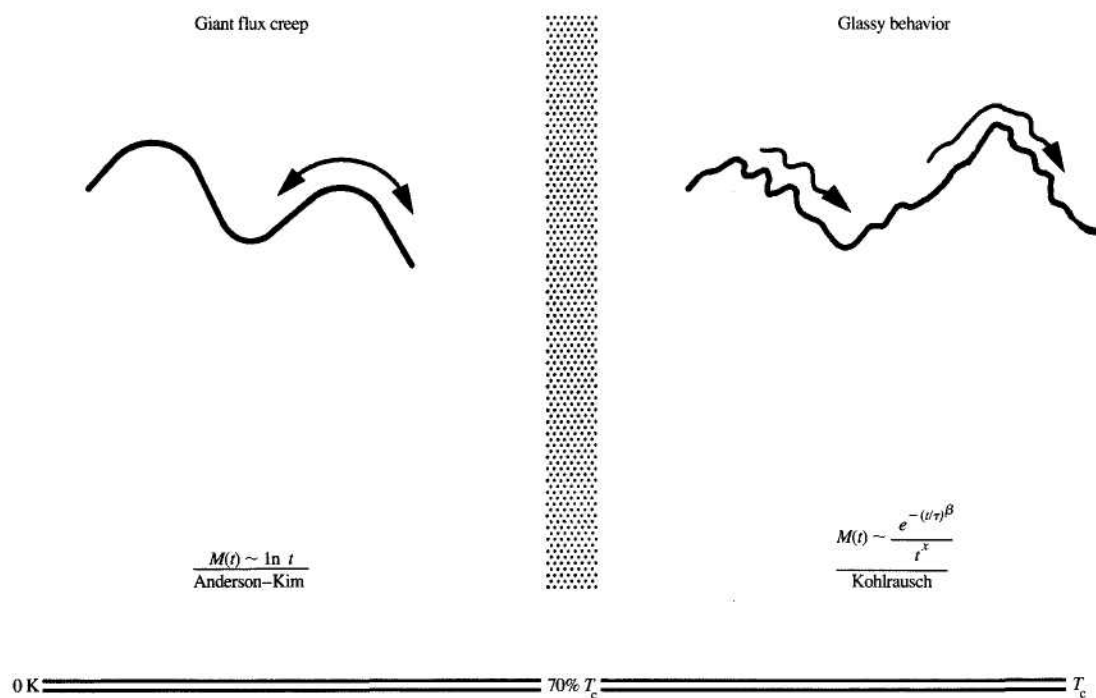


Figure 9

Schematic view of experimental situation. From [21].

that the short-time behavior of $M(t)$ can be very well fitted to $\ln t$. Therefore, only very time-consuming experiments can validate Equation (4), because knowledge of the entire decay is necessary to show Kohlrausch behavior unambiguously. Still, Kohlrausch decay does not prove glassy behavior, as it also exists in an Ising ferromagnet below T_c . It could, however, disprove it. Therefore, we have to look for better, more easily accessible experiments.

2. Aging

Rossel et al. [12] repeated the aging experiment, well known in spin glasses, for a superconducting single crystal. After the crystal was field-cooled in $H_0 = 0.05$ T and t_w aged up to 24 hours, the decay of the magnetization measured after switching the field by $\Delta H = 0.1$ T showed a characteristic shape shift just after the same time t_w . Such an effect can only be described by a hierarchy of barriers or the resulting broad distribution of relaxation times [12].

3. Time-dependent specific heat

This effect was found in simulations for particular spin-glass models [13]. It was not as strong as in recent experiments by A. Voronel [14]. This shows that the hierarchy in the high- T_c glasses is even more pronounced than in spin glasses. Furthermore, the strong effect can also be described in terms of a super-glass—a system with annealed and quenched disorder at the same time. In terms of our XY model, it means that the A_{ij} phase factors relax additionally on a time scale which is comparable to that of the whole system. Physically, we deal with structural transitions leading to a relaxation of the twins, microtwins, etc. in the system. That means that our weak links change with time, but they do not disappear. Disorder relaxes to a certain degree of weaker disorder. The resulting large energy fluctuations can explain the strong relaxation effect in the specific heat. We want to emphasize that the super-glass concept is currently being refined and will be published elsewhere. Especially for

experimentalists, it will open a new world of "super-glassy" effects to be explored.

4. Quasi-de-Almeida-Thouless line

Since this line is found only in the "glass regime," its origin is clearly related to the hierarchy of barriers. Other approaches, which take only the flux creep single-barrier picture into account, have to fail. At this point, it becomes clear why the certainly "oversimplified" XY spin-glass model was so successful. It contains the essential physics, the hierarchy of barriers, and therefore reproduces the (universal) $H^{2/3}$ behavior. The flux creep approach and the XY model each describe different sides of the same coin. The XY model can be considered a rather basic approach to the Landau-Ginzburg theory, one which just keeps the essential physics for a more qualitative description of the glass phase. Further intensive work is certainly necessary to obtain a quantitative theory for the glassy behavior.

5. Decoration experiment

A major point of criticism against a glass picture was the occurrence of an Abrikosov vortex lattice in single-crystal decoration experiments at Bell Laboratories [15]. But according to the glass picture, and in the spirit of small barriers experienced at lower temperatures, we should also expect to observe this phenomenon. At higher temperatures, when the hierarchy dominates, the effect should disappear. This was actually the case in the experiment which showed an Abrikosov lattice with a relatively short correlation length, at low T and small H . This situation was reproduced by T. Schneider and R. Hetzel in their approach to the XY model [16].

Conclusion

Summarizing at this point, we want to emphasize that a whole new world of glassy effects is waiting to be discovered in high- T_c experiments. It should be noted that in applications where we deal with nitrogen temperature, glassy behavior becomes dominant and must be understood. Especially when higher critical currents are reached in (ceramic) superconductors, glassy effects obviously play an important role. The current analysis of magnetization measurements may lead to critical currents which are much too low at higher temperatures.

From the theoretical point of view, our current situation is the following. We do not have a real contradiction between the (giant) flux creep and the glass picture. Flux creep is just a phenomenological macroscopic approach to the glassy behavior for low temperatures and relatively short times. Glassy behavior at higher temperatures and for relatively longer times may also be understood in terms of the following more phenomenological picture.

As already pointed out, the XY spin-glass model has to be considered a basic approach to the Landau-Ginzburg theory

including disorder. The XY spins are related to the different phases in the theory, and not to any physical spins. The term "spin-glass" should thus be understood with the emphasis on glass. Experimental results should be clearly separated from the Cu^{2+} spin glass, which occurs at much lower temperatures [17].

To visualize the physical situation according to the XY model is rather complicated. Therefore, we prefer a description in terms of flux or vortex lines (in a 3D system). In this sense we consider a certain type of "flux-line glass" to be just like a polymer glass in statistical physics, but with the polymers (flux lines) pointing in the direction of the magnetic field. The main point is that we have cooperative and therefore frustrated effects as we have (a) random pinning potentials and (b) interaction between different flux lines. Aspect (b) is different from the old superconductors, where for the flux creep theory only an isolated flux line—or, in the same spirit, a bundle of flux lines—was considered in a potential U , as mentioned above. Malozemoff et al. [18] generalized this picture, starting with a single barrier U , to a hierarchy of barriers. At this point it is an academic question whether we want to talk about glassy behavior. But we want to go a step further and consider not just one single flux line, but the cooperative behavior of an ensemble of flux lines. For high- T_c superconductors, the cutoff for the repulsive interaction between flux lines is much larger than in the case of traditional superconductors, owing to the short coherence length [19]. This means that at higher fields and higher temperatures we must include this interaction and the resulting frustration due to the random-pinning potentials. Pinning is therefore a cooperative effect. As with the traffic on a highway, a small disturbance (change of local pinning potentials) may easily lead to a traffic jam, in our case resulting in a higher critical current density j_c .

The picture outlined is currently under investigation in numerical simulations. Recent theoretical work by Matthew Fisher [20] supports the above picture, and we consider his "hard-core-boson world lines" to be equivalent to our flux lines. This derivation from Ginzburg-Landau theory shows the equivalence of the flux-line glass and the XY spin-glass pictures [20]. In general, however, we wish to emphasize that for the technically interesting temperatures around 77 K, critical currents are influenced by cooperative (glassy) pinning effects, in contrast to the traditional picture. For technical applications where higher j_c values are required, the cooperative (glassy) behavior is therefore of great importance.

References

- (a) I. Morgenstern et al., *Physica C* **153-155**, 59 (1988);
(b) K. A. Müller, M. Takashige, and J. G. Bednorz, *Phys. Rev. Lett.* **58**, 1143 (1987).
- For a recent review, see *Lecture Notes in Physics* **275**, Heidelberg Colloquium on Glassy Dynamics, J. L. van Hemmen and I. Morgenstern, Eds., Springer-Verlag, Berlin, 1987.

3. For a recent review on spin glasses in particular, see K. Binder and A. P. Young, *Rev. Mod. Phys.* **58**, 801 (1986).
4. J. R. L. de Almeida and D. J. Thouless, *J. Phys. A* **11**, 983 (1978).
5. I. Morgenstern, K. A. Müller, and J. G. Bednorz, *Z. Phys. B* **69**, 33 (1987).
6. C. Ebner and D. Stroud, *Phys. Rev. B* **31**, 165 (1985).
7. H. Keller, B. Pümpin, W. Kündig, W. Odermatt, B. D. Patterson, J. W. Schneider, H. Simmler, S. Connell, K. A. Müller, J. G. Bednorz, K. W. Blazey, I. Morgenstern, C. Rossel, and I. M. Savic, *Physica C* **153**, 71 (1988).
8. G. Deutscher and K. A. Müller, *Phys. Rev. Lett.* **59**, 1745 (1987).
9. A. C. Mota, A. Pollini, P. Visani, K. A. Müller, and J. G. Bednorz, *Physica C* **153**, 67 (1988).
10. L. Krusin-Elbaum, A. P. Malozemoff, Y. Yeshurun, D. C. Cronemeyer, and F. Holtzberg, *Physica C* **153**, 1469 (1988) and references therein.
11. See for example M. Tinkham, *Introduction to Superconductivity*, McGraw-Hill Book Co., Inc., New York, 1975.
12. C. P. Rossel, Y. Maeno, and F. H. Holtzberg, *IBM J. Res. Develop.* **33**, 328 (1989, this issue).
13. I. Morgenstern, in *Lecture Notes in Physics* **192**, Heidelberg Colloquium on Spin Glasses, J. L. van Hemmen and I. Morgenstern, Eds., Springer-Verlag, Berlin, 1983.
14. A. Voronel, private communication.
15. P. L. Gammel, D. J. Bishop, G. J. Dolan, J. R. Kwo, C. A. Murray, L. F. Schneemeyer, and J. W. Waszczak, *Phys. Rev. Lett.* **59**, 2592 (1987).
16. T. Schneider and R. Hetzel, to be published.
17. A. Aharony, R. J. Birgeneau, and M. A. Kastner, *IBM J. Res. Develop.* **33**, 287 (1989, this issue).
18. A. P. Malozemoff, T. K. Worthington, R. M. Yandrowski, and Y. Yeshurun, in *Towards the Theoretical Understanding of High Temperature Superconductors, Proceedings of the Adriatic Research Conference and Workshop, ICTP, Trieste, Italy*, S. Lundqvist, E. Tosatti, M. Tosi, and Yu Lu, Eds., World Scientific Press, Singapore, 1988.
19. G. Deutscher, private communication.
20. M. P. A. Fisher, preprint.
21. I. Morgenstern, K. A. Müller, and J. G. Bednorz, in *Proceedings of the 2nd Yukawa International Seminar (YKIS '88)*, Springer-Verlag, Berlin, in press.

I. Morgenstern IBM Research Division, Zurich Research Laboratory, Säumerstrasse 4, 8803 Rüschlikon/Zürich, Switzerland. Dr. Morgenstern studied physics and mathematics at the University of Saarbrücken from 1973 to 1978. He obtained his Ph.D. in physics in 1980 from the Kernforschungsanlage, Jülich. From 1980 to 1986, he was at the University of Heidelberg, where he obtained his Habilitation. Since 1987 Dr. Morgenstern has been on the research staff of the IBM Zurich Laboratory, where he is engaged in high- T_c superconductivity research.

Received December 23, 1988; accepted for publication January 10, 1989